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CONTAMINATION OF WATER DISCHARGE FROM CLINTON LABORATORIES

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CONTAMINATION OF WATER DISCHARGE FROM CLINTON LABORATORIES

by

A. Z. Morgan and Forrest Western

The radioactive waste products resulting from the operation of the Clinton Pile consist principally of water-borne fission products. The activity of this waste is reduced by storage to remove the short-lived products, precipitation of large fractions in underground tanks, precipitation in a settling basin, and adsorption on the clay of White Oak Creek and Lake. These processes are supplemented by dilution in White Oak Creek and Clinch River. Measurements of radiation and activity, together with fission product analyses, of the water discharged from White Oak Lake show that the average level of activity in White Oak Lake is kept well below tolerance values and that the activity in Clinch River is negligible.

Introduction

Four types of radioactive waste are, or have been, discharged from Clinton Laboratories, as follows:

1. Contaminated trash which is collected in special trucks and hauled to the "hot trash burial grounds." Here it is placed in deep holes or trenches and covered with several feet of dirt.
2. Radioactive argon discharged from the pile cooling air by way of the 105 stack.
3. Air borne fission products discharged from stacks into the atmosphere.

This type has included:

- a. Discharge from the 205 Plutonium Separations Plant via the 205 stack.
- b. Discharge from the 204 Plutonium Isolation Plant via the 205 stack.

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- c. Discharge from the 706C Fission Product Building and the 706D Barium Production Building via the 205 stack and the small 706C and 706D stacks.
 - d. Discharge from ruptured pile slugs via the 105 stack.
 - e. Discharge from the 706A Semiworks via the small 706A stack.
4. Water transported fission products, remaining in solution after partial precipitation, discharged into White Oak Creek. This has included:
- a. Discharge from the 205 Plutonium Separations Plant.
 - b. Discharge from the 706C and 706D Buildings.
 - c. Discharge from 706A Semiworks and Chemistry Buildings.
 - d. Discharge from the "hot" slug storage canal in the 105 Pile Building.
 - e. Discharge from the Tank Farm dry wells.

This discussion is concerned with the water transported fission products.

During the early operation of Clinton Laboratories, from December 1943 to January 1945, when the prime objective was to produce gram amounts of plutonium per day, most of the discharge of water-borne fission products was from the 205 Plutonium Separations Plant. Table 1 indicates the approximate activities of these fission products which contributed substantially to the radioactive contamination of discharged water. The activities given are for the products separated in an average daily run of the Separations Plant, at various times after removal from the pile; and are computed on the assumption that the batch was in the pile for 100 days, during which time it developed 1/100 of the pile power level of one megawatt. Before partial precipitation in storage tanks, all of these products were contained in the discharged water wastes except about 50% of the iodine

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which was deposited in the stack and off gas lines or was discharged into the air with the xenon. The uranium slugs were usually allowed to cool 40 days between removal from the pile and processing. During this time the elements with short half lives had decayed to negligible values, leaving a total activity, as indicated in the table, of about 1500 curies per day in the discharged water waste.

Table 2 gives the daily tolerance concentrations of the long-lived fission products in drinking water and for continuous submersion, respectively; together with data upon which the computations are based. The methods of computation are described in the paper, Tolerance Concentrations of Radioactive Substances ⁽¹⁾, and exposure of several years' duration is assumed. For all organs of the body except the thyroid, it is assumed that the tolerance level of radiation is 0.1 r per 24 hours, and that all of the energy of radiation liberated in the organ is absorbed in the organ. For the thyroid, on the basis of relative insensitivity of its tissue to radiation and of the small size of the organ, it is assumed that the tolerance level is 1 r per 24 hours and that only the energy of the beta radiation is absorbed.

Table 3 gives the comparative hazards to life that would result from the daily discharge of the entire activities of these fission products into the drainage system of the area, at the ages shown. The figures given are obtained by dividing the values given in Table 1 by the tolerance concentrations given in Table 2.

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TABLE I

Activities of Long Life Fission Products from Daily Separations Run

Activities are given in curies, and time is measured from removal of uranium from pile. Basis of computation is given on Page 2. Computed values contain, at most, two significant figures.

| | $\frac{\% \text{ Fission}}{\text{Yield}}$ | Half Life | Exp. (hr) | 40 Days | 100 Days | 365 Days | 730 Days |
|---|---|-------------|-----------|---------|----------|----------|----------|
| ^{89}Sr 1.5 | 4.6 | 53 days | 286 | 170 | 77 | 2.4 | 0.02 |
| ^{90}Sr | 5 | 25 years | 3.3 | 3.2 | 3.2 | 3.2 | 3.1 |
| ^{90}Y | (Sr ⁹⁰) | 60 hours | 3.1 | 3.2 | 3.2 | 3.2 | 3.1 |
| ^{91}Zr 1.4 | 5.9 | 57 days | 354 | 218 | 105 | 4.2 | 0.05 |
| ^{95}Zr 0.5 | 6.4 | 65 days | 358 | 234 | 165 | 7.3 | 0.15 |
| ^{95}Nb 0.5 | (Zr ⁹⁵) | 35 days | 210 | 240 | 179 | 14.5 | 0.30 |
| ^{103}Zr { 2.2 (99%) 0.8 (5%) | 3.7 | 42 days | 255 | 132 | 49 | 0.6 | 0.0015 |
| ^{106}Zr | 0.5 | 1 year | 7.4 | 6.9 | 6.1 | 3.7 | 1.8 |
| ^{127}I | 0.033 | 90 days | 1.5 | 1.1 | 0.7 | 0.09 | 0.005 |
| ^{129}I | 0.19 | 32 days | 14 | 6.0 | 1.6 | 0.005 | 0.000 |
| ^{132}I | 3.6 | 77 hours | 307 | 0.05 | 0.000 | 0.000 | 0.000 |
| ^{131}I | 2.8 | 8 days | 239 | 8.9 | 0.05 | 0.000 | 0.000 |
| ^{135}I | 5.6 | 20000 years | 0.5 | 0.5 | 0.5 | 0.5 | 0.5 |
| ^{137}I | 6 | 33 years | 2.9 | 2.9 | 2.9 | 2.9 | 2.8 |
| ^{140}Ba | 6.1 | 12.8 days | 520 | 59 | 2.3 | 0.000 | 0.000 |
| ^{140}Ba | (Ba ¹⁴⁰) | 40 hours | 520 | 68 | 2.6 | 0.000 | 0.000 |
| ^{141}Ce 0.4 | 5.7 | 28 days | 445 | 166 | 37 | 0.05 | 0.000 |
| ^{144}Ce 0.4 | 5.3 | 275 days | 100 | 91 | 78 | 40 | 16 |
| ^{143}Pr | 5.4 | 13.8 days | 458 | 61 | 3 | 0.000 | |
| ^{144}Ce 2.5 | (Ce ¹⁴⁴) | 17 minutes | 101 | 91 | 78 | 40 | 16 |

165
165
179
49
78
576

TABLE 2

Computation of Tolerance Values for Ingestion and Submersion

| Isotope | % Abs. in Critical Tissues | Critical Tissue | T _e Days | T _r Days | T Days | E Mev | Ingestion Tolerance m C/L | Submersion Tolerance m C/L |
|-----------------------------------|---|--------------------|------------------------|------------------------|-----------|----------|---------------------------------|----------------------------------|
| ⁸⁹ | 7.5 | Bone | 200 | 53 | 42 | 0.6 | 4.0 | 2.7 |
| ⁹⁰ -Y ⁹⁰ | 7.5 | Bone | 200 | 9125 | 196 | 0.9 | *0.58 | |
| ⁹⁰ | | | | | | 0.2 | | 8.1 |
| ⁹⁰ | 0.14 | Bone | 500 | 2.5 | 2.5 | 0.7 | 3100 | 2.3 |
| ⁹¹ | 0.14 | Bone | 500 | 57 | 51 | 0.5 | 210 | 3.2 |
| ⁹⁵ -Cu ⁹⁵ | 0.005 | Bone | 80 | 65 | 36 | 2.6 | *1600 | |
| ⁹⁵ | | | | | | 2.1 | | 0.77 |
| ⁹⁵ | 0.2 | Bone | 50 | 35 | 21 | 0.8 | 240 | 2.0 |
| ¹⁰³ | 0.00015 | Kidney | 20 | 42 | 14 | 0.8 | 11,000 | 2.0 |
| ¹⁰⁶ -Rh ¹⁰⁶ | 0.00015 | Kidney | 20 | 365 | 19 | 2.0 | *3200 | |
| ¹⁰⁶ | | | | | | 0.01 | | 160 |
| ¹⁰⁶ | The Half-life is so short that Rh ¹⁰⁶ has no effect except where it is in equilibrium with Ru ¹⁰⁶ | | | | | | | |
| ¹²⁷ | 1.5 | Kidney | 15 | 90 | 13 | 0.23 | 4.0 | 7.0 |
| ¹²⁹ | 1.5 | Kidney | 15 | 32 | 10 | 1.4 | 0.87 | 1.2 |
| ¹³² -I ¹³² | 1.5 | Kidney | 15 | 3.2 | 2.6 | 1.7 | *2.7 | |
| ¹³² | | | | | | 0.31 | | 5.2 |

(Table 2 continued on Page 7)

TABLE 2 - Continued

Computation of Tolerance Values for Ingestion and Submersion

| Isotope | % Abs. in Critical Tissues | Critical Tissue | T _e Days | T _{1/2} Days | T Days | E Mev | Ingestion Tolerance μ C/L | Submersion Tolerance μ C/L |
|--------------------|--|-----------------|---------------------|-----------------------|--------|-------|-------------------------------|--------------------------------|
| ¹³² I | 20 | Thyroid | 30 | 0.1 | 0.1 | *1.3 | 14 | 0.85 |
| ¹³¹ I | 20 | Thyroid | 30 | 8.0 | 6.3 | *0.57 | 0.55 | 2.8 |
| ¹³⁵ S | Activity is negligible and particle energies have not been determined. | | | | | | | |
| ¹³⁷ S | 30 | Muscle | 10 | 12000 | 10 | 0.97 | 5.4 | 1.7 |
| ¹⁴⁰ -La | 6 | Bone | 200 | 13 | 12 | 2.76 | *3.7 | |
| ¹⁴⁰ Ba | | | | | | 0.86 | | 1.9 |
| ¹⁴⁰ La | 0.11 | Bone | 16 | 1.7 | 1.5 | 2.4 | 1900 | 0.67 |
| ¹⁴¹ La | 0.18 | Bone | 50 | 28 | 18 | 0.42 | 560 | 3.8 |
| ¹⁴⁴ -Pr | 0.18 | Bone | 50 | 275 | 42 | 1.28 | 80 | |
| ¹⁴⁴ La | | | | | | 0.12 | | 13 |
| ¹⁴⁴ La | 0.002 | Bone | 40 | 0.01 | 0.01 | 1.16 | 3.3×10^7 | 1.4 |
| ¹⁴³ La | 0.002 | Bone | 40 | 13.8 | 10 | 0.3 | 1.3×10^5 | 5.4 |

*It is assumed here that only the parent isotope is ingested; the contribution of the daughter isotope is due to that amount formed in the body by disintegration of the parent.

**For computation of Ingestion Tolerance for Iodine, only average beta energies are used. These are: ¹³¹I, 0.2 Mev; ¹³²I, 0.5 Mev.

TABLE 2

Ratios of Activities from Table I to Tolerance Values for Ingestion and Submersion

| Isotope | 0 Days | | 40 Days | | 100 Days | | 365 Days | | 730 Days | |
|-------------------|-----------|------------|-----------|------------|-----------|------------|-----------|------------|-----------|------------|
| | Ingestion | Submersion | Ingestion | Submersion | Ingestion | Submersion | Ingestion | Submersion | Ingestion | Submersion |
| ⁹⁰ Y | 72 | 106 | 43 | 63 | 19 | 29 | 0.60 | 0.90 | 0.005 | 0.007 |
| | 5.7 | 0.41 | 5.5 | 0.40 | 5.5 | 0.40 | 5.5 | 0.40 | 5.3 | 0.38 |
| | 0.0010 | 1.3 | 0.0010 | 1.4 | 0.0010 | 1.4 | 0.0010 | 1.4 | 0.0010 | 1.3 |
| ⁹⁵ Cb | 1.7 | 111 | 1.0 | 68 | 0.50 | 33 | 0.02 | 1.3 | 0.0002 | 0.016 |
| | 0.22 | 465 | 0.14 | 304 | 0.10 | 214 | 0.0045 | 9.5 | 0.0001 | 0.20 |
| | 0.88 | 105 | 1.0 | 120 | 0.75 | 90 | 0.060 | 7.3 | 0.0013 | 0.15 |
| ¹⁰³ Pb | 0.023 | 128 | 0.012 | 66 | 0.0044 | 25 | 0.0001 | 0.30 | 0.0006 | 0.0008 |
| | 0.0023 | 0.046 | 0.0022 | 0.043 | 0.0019 | 0.038 | 0.0012 | 0.023 | 0.0006 | 0.011 |
| ¹⁰⁶ Rh | 0.38 | 0.21 | 0.28 | 0.16 | 0.18 | 0.10 | 0.023 | 0.013 | 0.0013 | 0.0007 |
| | 16 | 12 | 6.9 | 5.0 | 1.8 | 1.3 | 0.0057 | 0.0042 | 0.0000 | 0.0000 |
| ¹³² I | 114 | 59 | 0.019 | 0.0096 | 0.0000 | 0.0000 | 0.0000 | 0.0000 | 0.0000 | 0.0000 |
| | 435 | 85 | 16 | 3.2 | 0.09 | 0.018 | 0.0000 | 0.0000 | 0.0010 | 0.0000 |
| ¹³⁷ Cs | 0.54 | 1.7 | 0.54 | 1.7 | 0.54 | 1.7 | 0.54 | 1.7 | 0.52 | 1.6 |
| ¹⁴⁰ La | 140 | 274 | 16 | 31 | 0.62 | 1.2 | 0.0000 | 0.0000 | 0.0000 | 0.0000 |
| | 0.27 | 776 | 0.036 | 101 | 0.0014 | 3.9 | 0.0000 | 0.0000 | 0.0000 | 0.0000 |
| ¹⁴¹ Pr | 0.79 | 117 | 0.30 | 44 | 0.066 | 9.7 | 0.0001 | 0.013 | 0.0000 | 0.0000 |
| ¹⁴⁴ Pr | 1.3 | 7.7 | 1.1 | 7.0 | 0.98 | 6.0 | 0.50 | 3.1 | 0.20 | 1.2 |
| | 72 | 72 | | 65 | | 56 | 29 | 0.0000 | 0.0000 | 11 |
| ¹⁴³ Sm | 0.0035 | 85 | 0.0005 | 11 | 0.0000 | 0.56 | 0.0000 | 0.0000 | 0.0000 | 0.0000 |

Values for ingestion depend upon both mother and daughter, while values for submersion depend only on mother


A. Facilities for the Hold-up and Decontamination of Effluent Wastes

Arrangement of facilities for disposal of the chemical wastes from the Separations Plant and the other chemical units is shown in Figure 1. The wastes from Separations included aluminum nitrate solution resulting from dissolving of the aluminum coatings from uranium slugs, bismuth phosphate by-product precipitate, waste supernatants from the bismuth phosphate and Lanthanum Fluoride product precipitates, and various other wastes, in addition to fission products. These were stored in the large underground tanks, W5 and W6, where they were treated with sufficient sodium carbonate to raise their pH to about 8. This produced precipitation of radioactive materials, leaving only about 7% of the original radioactivity in the supernatant. Activity of the supernatant was further reduced by as long a hold-up for radioactive decay as storage space would permit.

It was originally planned that the two holding ponds, of about 225,000 gallons capacity each, would alternately receive the supernatant from tanks W5 and W6 together with the contaminated water from the rest of the plant. The water in the east pond was to "cool" while the west pond was being filled. Then the east pond would be drained into White Oak Creek and filled while the west pond cooled. Discharge into these ponds did not begin until March 6, 1944; and it was discontinued April 27, 1944, because of the large amount of fission products measured in the water and mud of the White Oak drainage system during the latter part of April. (2)

Inadequacy of the ponds was largely due to inadequate size and to the fact that much of the precipitate that settled in the ponds washed out into White Oak Creek when the ponds were drained. At present these ponds are not used except for infrequent short intervals during which it is desirable to divert waste water from the Settling Basin.

While the holding ponds were being used, a calcium precipitate was observed to be collecting at their outlet and for several hundred yards



LEGEND
 * VALVE
 □ DIVERSION BOX PLUG VALVE
 + BLANK IN LINE
 ○ MANHOLE
 HPW BY PROD WASTE
 BO BOTTOM OUTLET
 OF OVERFLOW
 J JET

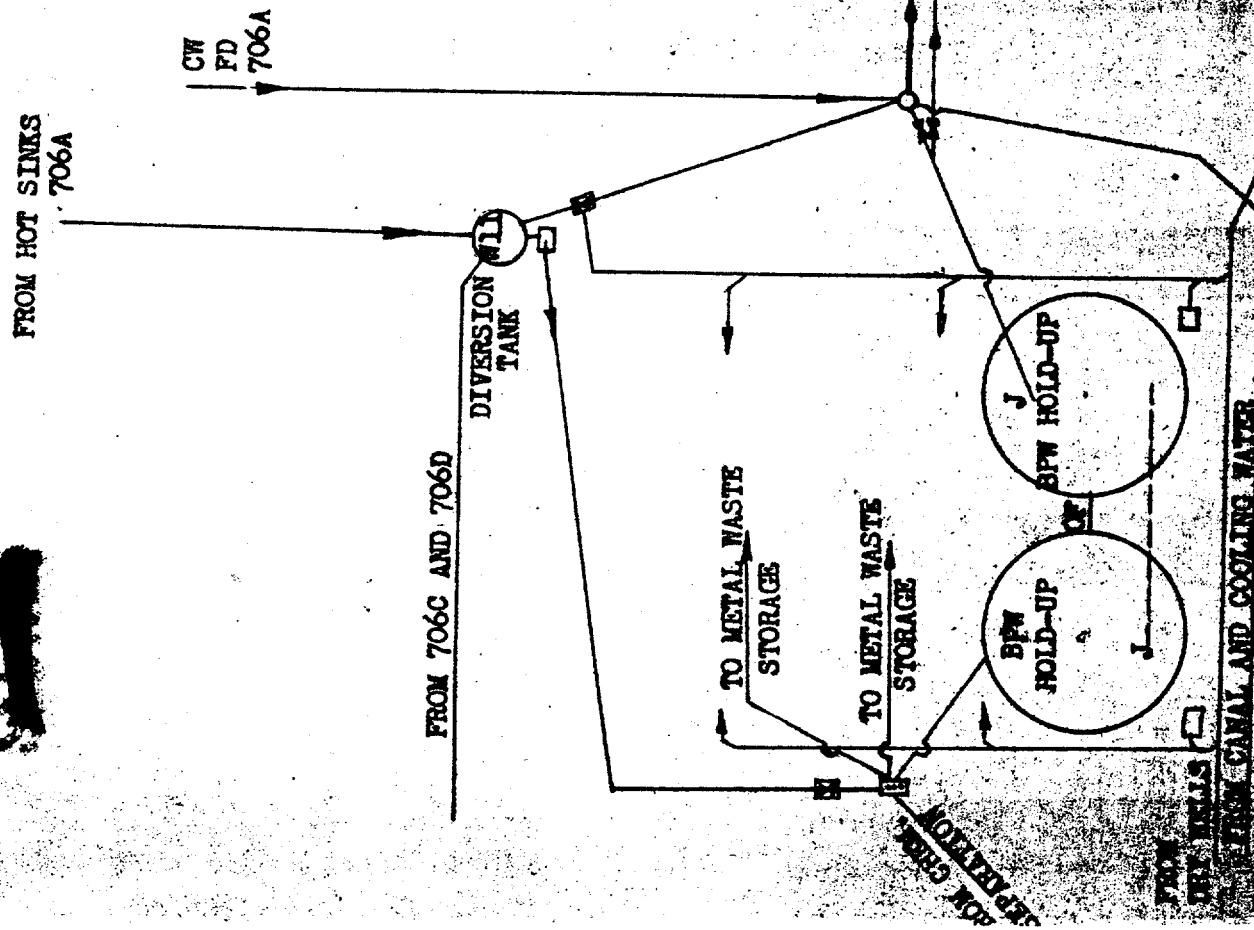


FIGURE 1

down White Oak Creek. This suggested the addition of calcium chloride to the water in tanks W5 and W6. This treatment, which was used from April 17 until discharge into the pond was discontinued on April 27, 1944, reduced the activity of the waste water about 50%.

As a consequence of the inadequacy of the holding ponds, a 1,600,000 gallon settling basin was built, and put into operation July 3, 1944. Experiments had indicated that dilution of the supernatant from W5 and W6, in the ratio of 70 to 1, would precipitate about 90% of the remaining radioactive contamination. The discharge from these tanks is mixed with the dilution water of the plant to give a flow of about 900,000 gallons of water per day and is fed into the settling pond through a diversion box and a weir which distributed it rather evenly on the north side of the basin by means of a number of discharge slots. Radioactive material precipitated by this dilution has to be cleaned from the inlet weir occasionally to prevent excessive radiation along the catwalks across the basin. A quantitative record of the effectiveness of the settling pond in removing radioactive material from the discharged waste is summarized in Table 5.


When the settling basin was constructed, it was considered desirable to construct a series of baffles over the basin to increase the length of path of water flow and to insure a maximum of settling. However, there was not sufficient time for the most careful design and construction; and, as a compromise, a wooden framework was floated on top of the water to prevent surface water from blowing directly along the water surface to the outlet, and to assist in dispensing the undersurface water. A weir on the south side of the basin receives the effluent water from many slots and feeds it into White Oak Creek.

On July 10, a week after the discharge of waste into the new settling basin began, the procedure of putting 500 pounds of calcium chloride per day into tank W5 was resumed. This treatment reduced the activity of the waste

by a slurry of about 1000. Treatment of the water in the settling basin with similar amounts of calcium chloride was tried for about three weeks but was discontinued as ineffective.

During the summer of 1944, various forms of algae began to grow luxuriantly on the bottom of the basin, on weir boxes, and on the baffle system in the basin. When carbon dioxide collected in the spongy algae, they floated on the water surface and formed a green carpet over almost the entire basin. These algae contained various radioactive fission products fixed in and on the surface of the plant cells. The resulting radiation level was frequently several hundreded milliroentgens per hour along the catwalks and of the order of one roentgen per hour just above the algae-covered basin surface. Baffles were constructed about the outlet weir, but, in spite of all efforts, bits of "hot" algae continued to escape into White Oak Creek. It was observed that after rains the algae settled to the bottom of the basin for about a day, so fire hoses were played frequently on the water surface to simulate heavy rain. Finally, in November of 1944, on the recommendation of R. E. Zirkle, 200 gallons of 37% formaldehyde were added to the basin water and allowed to stand without any flow in or out of the basin for three days. This treatment plus the winter weather killed the algae completely. Small amounts of algae collected in the basin during the summers of 1945 and 1946, but failed to present a serious problem.

On September 19, 1944, there was a cloud-burst of 8.8 inches of rain. White Oak Creek filled very rapidly and overflowed the settling basin for two hours at the rate of about 1000 gallons per minute. No serious damage was done to the basin, and the enormous dilution prevented development of any radiation hazard downstream. Some of the algae were temporarily cleared from the basin and, as a result, the radiation level above the catwalks was



removed by a factor of 20. A negligible fraction of the sediment was removed, however, from the bottom of the basin. The settling basin, as well as White Oak Dam, came very near washing away during this flash flood. To remove the threat of a future washout of the basin which might discharge a thousand or more curies of activity into the drainage system, the bed of White Oak Creek near the basin was widened and deepened.

Since the shutdown of the Chemical Separations Building (205), most of the "hot" waste comes from the Fission Product Building (706C) and the Barium Separations Building (706D) by way of the small tank W11. This tank overflowed in November, 1944, and so an automatic jet was installed to transfer fluid from W11 to W5. A large increase in the per cent of barium and strontium found in the fission products carried by the water of White Oak Lake in December, 1944, suggested that the jet from W5 and W6 might be picking up sludge from W5. As a consequence, this jet was replaced by an overflow line. The small retention pond, shown in Figure 1, connects to all of the dry wells about the tanks. Readings which are made of this water daily indicate when there is a spill or leak about the tanks.

B. The By-Products Waste Drainage System

The waste drainage system is shown in Figure 2. Some 1500 feet below the Settling Basin outlet, the flow of White Oak Creek is deflected from its natural course by a dike to form a marsh. A second dike provides some hold up in the intermediate pond and returns the water to its original course. White Oak Dam, two miles downstream, provides a lake which impounds 9,400,000 cubic feet of water when filled. The dam has an upper control gate which is used to regulate the flow, and the resultant dilution, of the water passing into the Clinch River; and a lower control gate which is opened only during extreme emergency. The partial removal of activity from the waste water by adsorption on the clay in the mud of this

WMSM

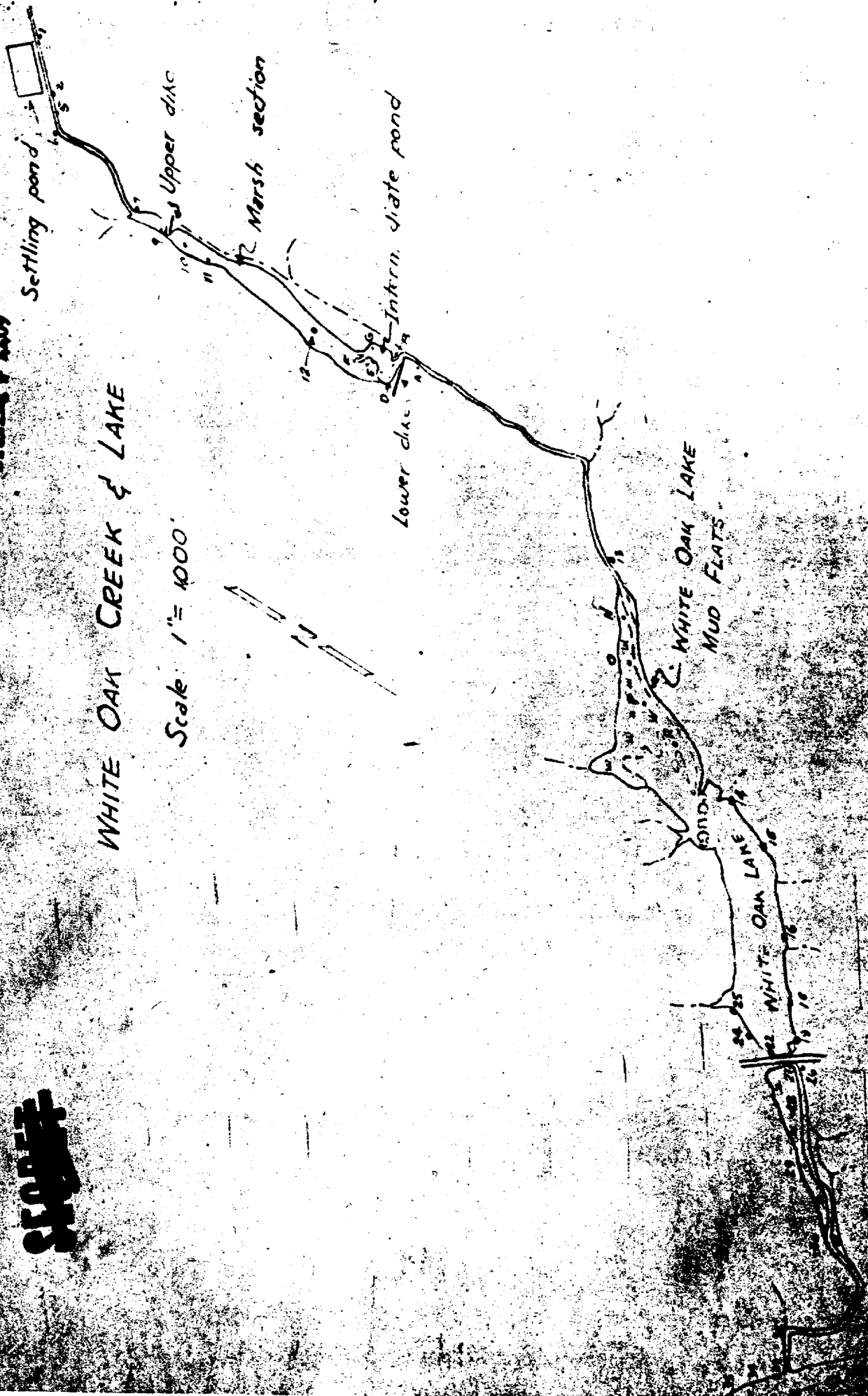
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Fig 2

Drawing # 4409

WHITE OAK CREEK & LAKE

Scale 1"=1000'



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system is discussed in detail in another paper (3) in this volume.

The flow of Clinch River is controlled by Norris Dam which is about 40 miles above the mouth of White Oak Creek. The average flow of Clinch River is given in Table 4. In case of emergency, arrangements can be made to increase the rate of flow for short periods of time in order to obtain greater dilution of the fission product waste.

TABLE 4

Average Flow of Clinch River

| | |
|-----------|---------------------------|
| 1921-1932 | 4600 ft ³ /sec |
| 1940 | 2200 |
| 1941 | 2000 |
| 1942 | 2500 |
| 1943 | 5190 |
| 1944 | 4610 |
| 1945 | 4800 |
| 1946 | 5000 |

5. Measurements of the Radioactivity of Discharged Water Wastes

It is the responsibility of the Health Physics Section to see that no concentration of radioactive products is discharged into White Oak Creek that would be harmful to man or animal. From the start of discharge of radioactive contamination into White Oak Creek, measurements of radioactivity have varied from time to time with the development of new instruments and techniques.

Operations personnel make daily measurements with a Lauritsen electroscope at various locations, formerly about the retention ponds, and, later, about the Settling Basin. They also collect samples from the affluent and effluent waters of the ponds and basin several times per day.

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These samples are evaporated and counted for beta activity by the 205 Analytical Section. (4,5) Monthly summaries of radioactive contamination estimated from these counts are given in Table 5.

The Health Physics Section makes regular Lauritsen electroscope measurements about the ponds and basin. Gamma ray measurements are made several times a week in large samples of water taken from the discharge of the Settling Basin and of White Oak Lake. Formerly, measurements were made occasionally by floating X-22 ionization chambers over the Basin and White Oak Lake, and with a submarine counter in White Oak Lake and Clinch River. More recently, continuously recording GM counters have kept records of the beta and gamma activity at White Oak Lake Dam. These counters have proven valuable for the early detection of changes in the activity of the water leaving the lake. Routine samples of water discharged from White Oak Lake are collected and analyzed or counted as discussed in the next section.

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TABLE 5

| Month 1944 | Settling Basin | | | | White Oak Lake | |
|---------------|----------------------|-----------------------|-------------------------|---------------------------------|---|---------------------------------|
| | Beta Curies In | Beta Curies Out | Beta Ratio In/Out | Average Gamma Out (mr/hr) | Beta Curies Out per ml $\times 10^{12}$ | Average Gamma Out (mr/hr) |
| August | 90 | 13 | 6.9 | | | |
| September | 61 | 11 | 5.5 | | 32 | |
| October | 251 | 35 | 7.2 | 0.75 | 115 | *0.050 |
| November | 277 | 74 | 3.7 | 1.24 | 108 | |
| December | 156 | 60 | 2.7 | 0.91 | 45 | |
| 1945 | | | | | | |
| January | 96 | 44 | 2.2 | 0.43 | 79 | 0.039 |
| February | | | | 0.01 | 9 | 0.029 |
| March | 6 | 4 | 1.7 | 0.07 | | 0.012 |
| April | 89 | 53 | 1.7 | 0.90 | | 0.012 |
| May | 66 | 40 | 1.7 | 0.24 | | 0.012 |
| June | 94 | 55 | 1.7 | 0.65 | | 0.041 |
| July | 57 | 38 | 1.5 | 0.24 | | 0.019 |
| August | 110 | 82 | 1.3 | 0.49 | | 0.035 |
| September | 104 | 93 | 1.1 | 0.53 | | 0.050 |
| October | 22 | 24 | 0.9 | 0.16 | 18 | 0.033 |
| November | 8 | 11 | 0.7 | 0.04 | 36 | 0.003 |
| December | 65 | 34 | 1.9 | 0.58 | 27 | 0.045 |
| 1946 | | | | | | |
| January | 207 | 173 | 1.2 | 1.19 | 18 | 0.141 |
| February | 39 | 23 | 1.7 | 0.028 | | 0.011 |
| March | 55 | 21 | 2.6 | 0.532 | | 0.049 |
| April | 73 | 65 | 1.1 | 0.633 | | 0.039 |
| May | 161 | 97 | 1.7 | 0.586 | | 0.078 |
| June | 55 | 7 | 1.2 | 0.218 | | 0.082 |
| July | 74 | 57 | 1.3 | 0.288 | | 0.045 |
| August | 7 | 4 | 1.7 | 0.022 | | 0.012 |
| September | 38 | 30 | 1.3 | 0.048 | | 0.021 |
| October | 17 | 17 | 1.0 | 0.050 | | 0.035 |
| November | 46 | 40 | 1.2 | | | 0.047 |
| December | 55 | 41 | 1.3 | 0.008 | | 0.014 |

*This is an average of infrequent observations made during the last three or four months of 1944. Data immediately following the spill from W-11 on November 22, are not included.

D. Fission Product Analyses

From September, 1944, to February, 1945, monthly determinations of the average distribution of fission products in the water leaving White Oak Lake were made. The analyses were made by the Analytical Section on a composite sample accumulated during the month. The method included chemical separation of the principal long life fission products, evaporation to dryness, and measurement of the beta activities of the separated elements. Results of these analyses are given in Table 6. The concentrations of individual elements were too low for accurate measurements, resulting in inconsistencies to be observed in the table. Since the concentrations of strontium and barium were much too low to constitute individual hazards from ingestion, routine monthly analyses were discontinued with the sample for January, 1945; and subsequent monthly samples have been counted for gross beta activity only.

If at any time there is a high gross count, a fission analysis of the water is made in order to determine the tolerance level. Constant vigilance must be maintained to make certain that isotopes readily absorbed by the human body are not concentrated in the discharged water to such an extent that ingestion tolerance is exceeded. Usually tolerance is determined by the level of water contamination that will irradiate a submerged body to the extent of 100 milliroentgens per day. However, the tolerance levels for I^{131} , Sr^{90} , Te^{127} , Te^{129} , and Te^{132} are determined by ingestion of these elements in drinking water. The tolerance concentrations of these elements in drinking water are, from Table 2: I^{131} , 5.5×10^{-10} curies/milliliter; $Sr^{90} - Y^{90}$, 5.8×10^{-10} curies/ml; Te^{127} , 4.0×10^{-9} curies/ml; Te^{129} , 8.7×10^{-10} curies/ml; $Te^{132} - I^{132}$, 2.7×10^{-9} curies/ml.

TABLE 6

Fission Product Analyses of Water from White Oak Dam

| Month | Barium | | | Strontium | | | Zirconium | | | Columbium | | | Totals | | | Gross | |
|-------|--------|----|-----|-----------|----|-----|-----------|-----|------|-----------|------|------|--------|-------|------|-------|---|
| | 2 | | | 3 | | | 4 | | | 5 | | | 6 | | | | |
| | C | %B | A | C | %B | A | C | %B | A | C | %B | A | C | %B | A | 1 | B |
| 1944 | | | | | | | | | | | | | | | | | |
| t | 0.1 | 3 | 0.3 | 0.2 | 6 | 0.7 | 0.03 | 0.9 | 0.15 | 0.4 | 1.1 | 9.7 | 0.73 | 21.4 | 10.9 | 3.5 | |
| | 0.8 | 6 | 3.1 | 2.0 | 16 | 7.1 | 4.0 | 31 | 20 | 3.2 | 25 | 74 | 10 | 78.4 | 104 | 12.8 | |
| | 2.1 | 18 | 8.2 | 3.7 | 31 | 13 | 4.7 | 39 | 24 | 1.8 | 15 | 42 | 12.3 | 103 | 86 | 12.0 | |
| v 27) | 2.0 | 15 | 7.8 | 3.0 | 23 | 11 | 2.0 | 15 | 10 | 6.0 | 46 | 138 | 13 | 100 | 166 | 13.0 | |
| | 1 | 20 | 3.9 | 3 | 60 | 11 | 1 | 20 | 5 | ----- | ---- | ---- | 5 | 1.0 | 20 | 5 | |
| 45 | 2.1 | 35 | 8.2 | 2.8 | 47 | 10 | 11 | 180 | 55 | 5 | 83 | 115 | 26 | 430.4 | 218 | 6 | |

1 Notes: C and B - Beta counts per minute per milliliter.

2 %B - Percentage of gross beta counts per minute per milliliter as determined from aliquot of original sample.

3 A - Activity in curies per milliliter $\times 10^{-12}$, computed assuming:

counter geometry, 12.5%; back scattering, 10%; and absorption by 10 mg/cm² of Al.

On the basis of these assumptions, the overall correction factors used were:

Barium, 8.7; Strontium, 7.9; Zirconium, 12.6; Columbium, 51.

4 Deviation of these values from 100% are indicative of difficulties encountered by Analytical Department in analysis of these low activities.

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As indicated in Table 1, because of differences in the half lives of these isotopes, the relative amounts to be found in water wastes depends upon the age of the waste. Table 3 shows that for very young wastes the isotopes producing the greatest ingestion hazards are, in order of magnitude of hazard: I^{131} , Ba^{140} , Te^{132} , Sr^{89} , and Te^{129} . As the age of the waste increases, these become unimportant relative to the long lived Sr^{90} in the order: Te^{132} , I^{131} , Ba^{140} , Te^{129} , and Sr^{89} .

Three analyses of samples of water leaving White Oak Lake during unusual conditions are given in Table 7. The first of these was taken during the flood of September 29, 1944; the second during a period of unprecedented activity, January 30, 1946, resulting from a number of unusual circumstances coincident with dissolving slugs more recently removed from the pile than normal; and the third during the month of March, 1946, covering a period of several weeks in which the activity in the Settling Basin was far above normal. The near tolerance level in White Oak Lake represented by the second of these analyses was an unusual event of short duration. To provide a margin of safety for such unusual conditions, the level of the lake is normally kept 3.5 feet below the top of the upper gate of the dam, so that whenever the activity of the water at the dam exceeds 10% of tolerance the upper gate may be closed to allow additional decay, settling, absorption, and dilution of the radioactive isotopes before discharge into the Clinch River. The reserve capacity of the lake at this level is 5,000,000 cubic feet. In the event of heavy precipitation at such a time, reduction of activity results principally from dilution. Further dilution of waste discharged from the dam by the water of Clinch River has always reduced the radiation level to less than 1% of tolerance at a short distance below the mouth of White Oak Creek.

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TABLE 7

Analyses of Water from White Oak Lake Dam
At Times of Unusual Activity

September 29, 1944

| <u>Constituent</u> | <u>Solution</u> | | <u>Suspended Material</u> | |
|--------------------|----------------------|-----------------------------|---------------------------|-----------------------------|
| | <u>Counts/min/ml</u> | <u>μc/ml</u> | <u>Counts/min/ml</u> | <u>μc/ml</u> |
| Barium | 0.04 | 0.16 | 0.04 | 0.16 |
| Strontium | 0.13 | 0.47 | 0.07 | 0.25 |
| Zirconium | 0.01 | 0.05 | 0.02 | 0.10 |
| Columbium | 0.06 | 1.38 | 0.36 | 2.28 |
| Total | 0.24 | 2.06 | 0.49 | 8.79 |
| Gross | 1.61 | | 1.90 | |

January 30, 1946

| <u>Constituent</u> | <u>Counts/min/ml</u> | <u>μc/ml</u> |
|--------------------|----------------------|-----------------------------|
| Strontium | 40 | 156 |
| Iodine | 54 | 238 |
| Total | 94 | 394 |
| Gross | 87 | |

Month of March, 1946

| <u>Constituent</u> | <u>Counts/min/ml</u> | <u>μc/ml</u> |
|--------------------|----------------------|-----------------------------|
| Strontium | 2 | 7 |
| Gross | 8 | |

The incident of January, 1946, illustrates the above method of handling unusual activities. On January 24th, delayed reports on the activity of water leaving the Settling Basin, and reports from GM counters at the dam, indicated that the activity of the lake might be 10 times its normal value. The upper gate of the dam was closed, jetting of waste into the settling basin was stopped, and much of the plant water was diverted from the settling basin into the holding ponds. A $1\frac{1}{2}$ " rain January 30th filled the lake, diluting the activity to about one half of its peak value. Additional rainfall made it necessary to discharge water over the spillway, but it had reduced the activity to approximately its normal value by February 4th. The sample for which the analysis is given in Table 7 was taken at the dam, January 30, before overflow started. On the following day, samples were taken from three different points downstream in the Clinch River, from which it was estimated that any activity present was less than the probable counting error in the analysis.

Measurements of the beta activities of water entering and leaving the Settling Basin are made from samples of water taken at intervals of four hours by the Technical Division. From these and from the daily flow the number of curies for each day is computed. Average monthly beta activity of water leaving White Oak Lake is estimated from a composite sample of water accumulated through the month by the Health Physics Group and measured by the radiochemistry analysis group of the Chemistry Division.

Gamma radiation in the water leaving the Settling Basin and leaving White Oak Lake is measured in samples taken on alternate days from the two sources to total five or six samples per week. The method of measurement and correction for the finite size of sample are given by K. Z. Morgan (5).

For comparison between the two methods of monitoring the water, measurements of the beta activity and the gamma radiation in the water discharged from the Settling Basin are shown graphically in Figure 3, for those days in 1945, for which observations of the gamma radiation were made. Although there are some discrepancies, the agreement between the beta and gamma measurements is quite good, and lends confidence to the use of gamma measurements as a convenient method of monitoring the water flowing from White Oak Lake. However, it must be remembered that the beta-gamma ratio varies with plant operations, and with physical and chemical conditions in the Settling Basin and drainage system. For this reason, one cannot depend upon a single type of measurement for an extended period of time; and final interpretation is dependent upon a knowledge of the distribution of fission products in the water.

E. Effectiveness of Settling Basin and White Oak Lake
in Reducing Radioactive Contamination

Early investigation indicated that activity due to fission products would be reduced by a factor of 13 by precipitation in W6, and by an additional factor of 10 in the Settling Basin. It will be noted from Table 5 that the actual reduction factor for the Settling Basin has decreased from seven to something less than two. The graphs in Figure 4 show daily values of beta activities entering and leaving the Settling Basin from August 14, 1944, to December 31, 1944, during which period the greatest change took place. It may be observed from these graphs that the change occurred rather suddenly, confirming the belief that the cause was the change in operating conditions.

Comparison of gamma radiation levels in the water leaving the Settling Basin and that leaving White Oak Lake is indicated graphically for the year 1945 in Figure 5. These data show the gamma radiation to be reduced by an average factor of 13. While perhaps less reliable, data

FIGURE 3

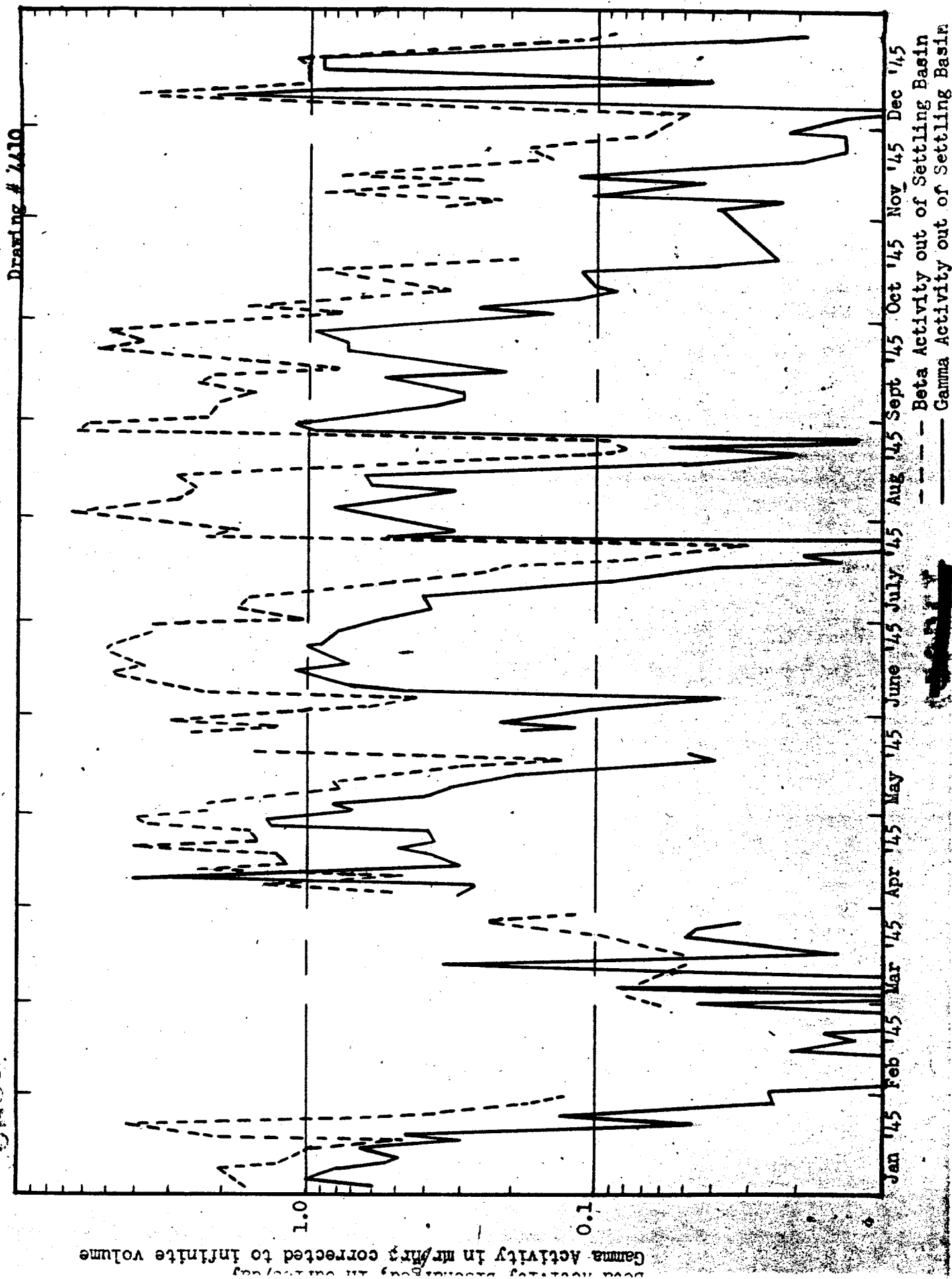
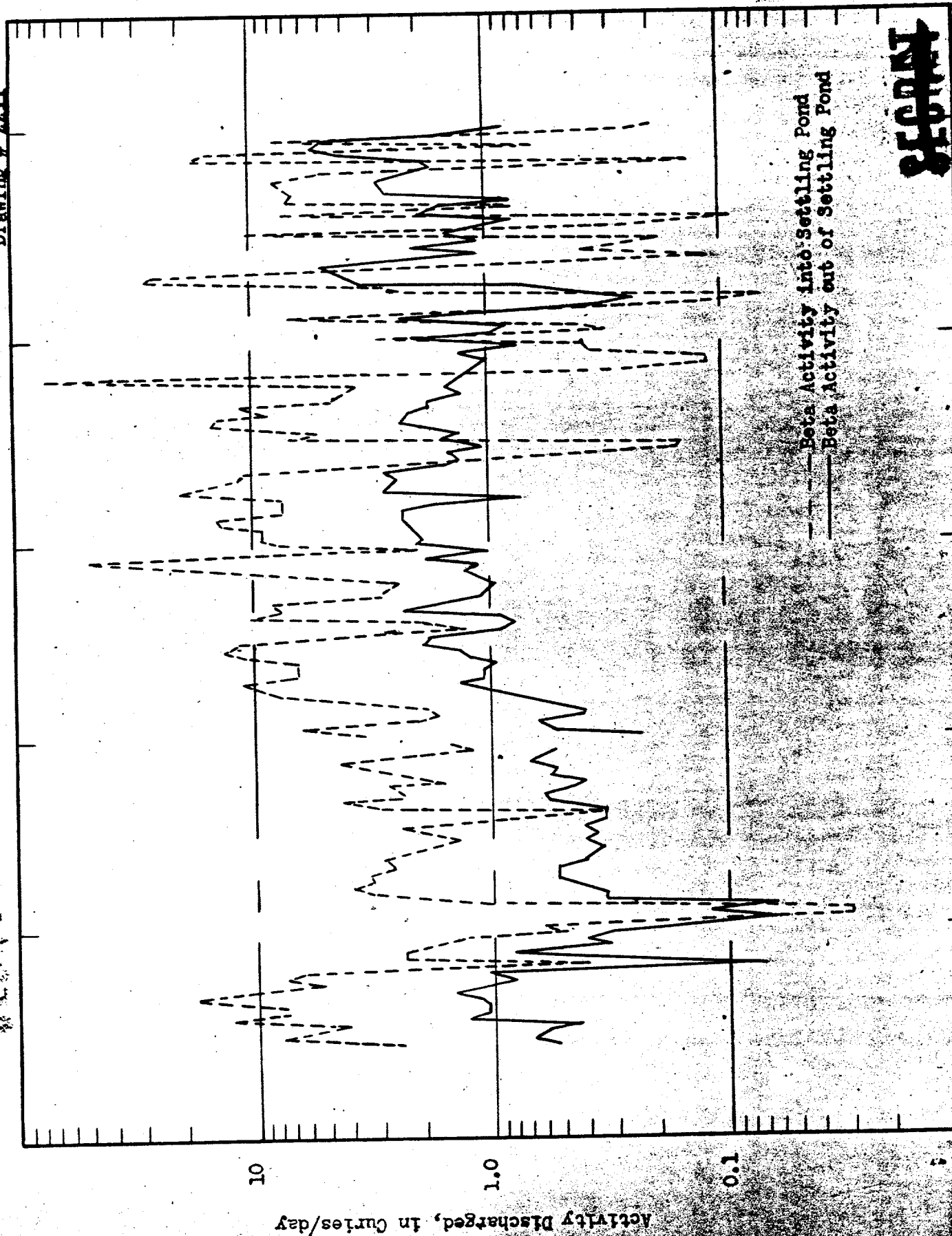


FIGURE 4

Drawing # 4411

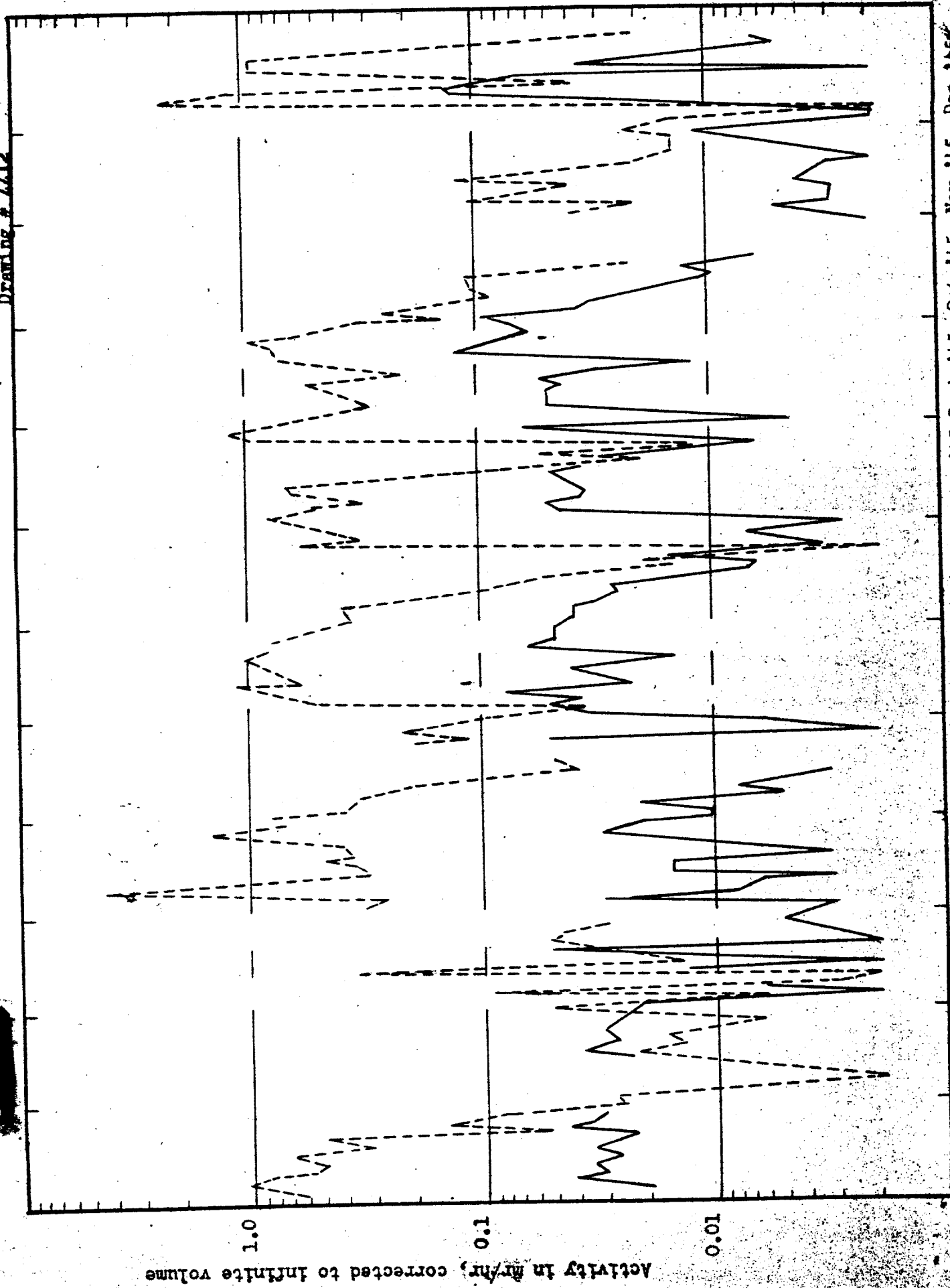


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FIGURE 5

Drawing # 4412



on beta activities, from Table 5, indicate a reduction in beta activity of the order of 10. The reduction is due to dilution, to settling, and to adsorption on the clay in the mud of the creek and lake. (3) When short lived radioisotopes, resulting from the processing of "young" slugs, are discharged from the plant, physical decay becomes an important factor.

F. Hazards to Life Resulting from Discharged Water-borne Wastes.

The plants and animals found in and about White Oak Creek and Lake, between the Settling Basin and Clinch River, appear to be those normal to the vicinity. Radiographs of plants gathered along the stream show that many of them are radioactive and, perhaps, are not desirable food for cattle. Studies of fish from the creek and lake, made by the Biology Section from May 3, 1944, to April 3, 1945, indicated a correlation between the activity of mud in the lake and beta activity in the tissues of the fish; but it was concluded that the radiation from the ingested radioactive wastes was well below tolerance for the fish, and that the concentrations of fission products in the tissues of the fish were below tolerance for human consumption. However, the water discharged from White Oak Lake passes through a fish screen which, except during periods of flood, prevents the passage of the radioactive fish into the Clinch River. Signs posted around the lake prohibit fishing, as well as swimming or use of the water for human consumption.

In the spring of 1945, the death of a large number of fish in White Oak Creek and Lake was investigated by the Biology Section, (6) but no connection with radioactive wastes was found. All of the dead fish were of one species, known to be very delicate, while the lake contains many other species. In the spring of 1946, the very young fish in the shallow water of White Oak Creek near the Settling Basin died and young fish were not observed in this portion of the creek for several months. While the circumstances of this incident were entirely different from the preceding one, there is no evidence that it resulted from the radioactive nature of the water

Possible damage to life from the radioactive waste discharges into the water may result either from external radiation or from the ingestion of radioactive products which become fixed in the organs of the body. The following table summarizes activities of discharged water during the period from August, 1944, to December 31, 1945:

TABLE 3

Beta and Gamma Activities of Discharged Water
For the Period August 1944 - December 1945.

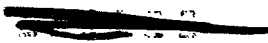
| | <u>From Settling Basin</u> | <u>From White Oak Lake</u> |
|--|---------------------------------|----------------------------|
| Average gamma radiation | 0.5 mr/hr | 0.027 mr/hr |
| Maximum gamma radiation | 1.2 mr/hr | 0.12 mr/hr |
| Average beta activity | 3.9×10^{-10} curies/ml | 5×10^{-11} c/ml |
| Maximum beta activity | 1.8×10^{-9} curies/ml | 2×10^{-10} c/ml |
| Maximum concentration of strontium plus barium | | 2.1×10^{-11} c/ml |

^aThese values represent the highest monthly average values for the period covered by this table. An incident in which much higher values were attained for a brief time is discussed on page 22.

From tolerance values for ingestion and submersion given in Table 2, it will be observed that the activities shown above are well within the tolerance limits for man. All measurements of the radioactivity of the water downstream in Clinch River indicate a level of radiation of the order of background radiation.

Present Status of the Problem of Disposal of Radioactive Wastes.

While it is believed that the methods of waste disposal used at Clinton Laboratories until the present time have met an emergency situation without endangering the health of personnel within or without the area of operation, it is now considered desirable by the authors of this report to modify the system as soon as it is technically practical to do so. Instead of diluting the small fraction of activity remaining in the supernatant after precipitation in such underground tanks as W5 and W6 and discharging it into a drainage system as is done now, the remaining activity should be concentrated by some physio-chemical process until it would be practical either to store it in additional underground tanks or to bury it in suitable, well marked and permanently cataloged burying grounds.



References

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- (3) J. S. Cheka and K. Z. Morgan: Radioactive Fission Product Contamination in the Mud of White Oak Drainage System. This Volume
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- (5) K. Z. Morgan: Operating Equations and Procedures Involved in Water Counting at Site. CH-2565. January, 1945
- (6) H. J. Curtis to R. L. Doan: Fish in White Oak Lake. April 23, 1945